Band Mapping in Higher-Energy X-Ray Photoemission: Phonon Effects and Comparison to One-Step Theory

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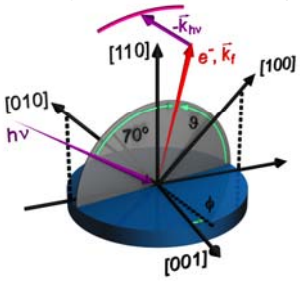
Introduction: There is growing interest in extending valence electronic studies with angle-resolved photoemission (ARPES) into the soft x-ray and hard x-ray regimes. In comparison to low-energy band mapping, however, additional effects are present. These include the increased angular resolution required to probe a small enough region in the Brillouin zone, the need to allow for the photon momentum in wave-vector conservation, and thermal effects due to phonon creation and annihilation during photoexcitation that smear out the specification of the initial wave vector, finally leading to a density of states spectrum.

Experimental geometry

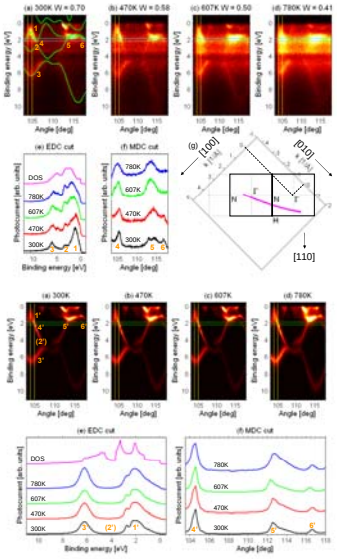


$$\vec{k}_f = \vec{k}_i + \vec{g}_{hk\ell} + \vec{k}_{hv} + \vec{g}_{surf} + \vec{q}_{phonon}$$

$$\vec{k}_i \approx \vec{k}_f - \vec{k}_{hv} - \vec{g}_{hk\ell}$$

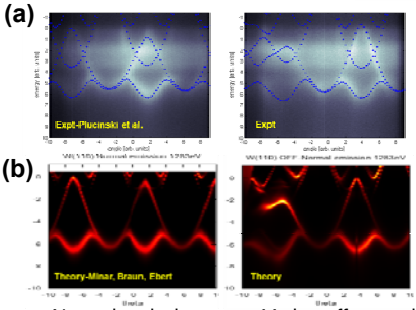
Experiment: Measurements were carried out on a (110)-oriented tungsten crystal at beamline 4.0.2 of the Advanced Light Source in Berkeley, using the Multi-Technique Spectrometer/Diffractometer located there.

870 eV Theory Experiment



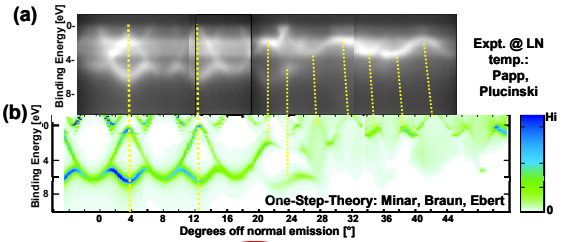
Results and discussion: We have analyzed the data in first approximation by requiring wave-vector conservation between bulk band states and ***free-electron final states***, with the *k*-conservation condition being modified to allow for the ***photon momentum***. Furthermore, we have compared the experimental data with state-of-the-art ***one-step photoemission theory*** [2] which includes a precise evaluation of matrix element effects and an attempt to include phonon effects via complex phase shifts. Good agreement is found as to peak positions and relative intensities, although ***phonon effects are not correctly predicted***, indicating a need for improvement in theory (see Ebert talk).

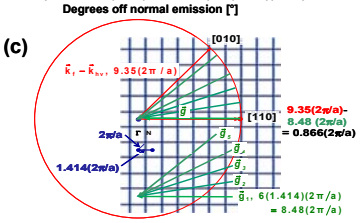
1253.6 eV-Mg Kα



Normal emission 11 deg off-normal

(a) Experimental detector images and free-electron final-state theory. (b) One-step theory

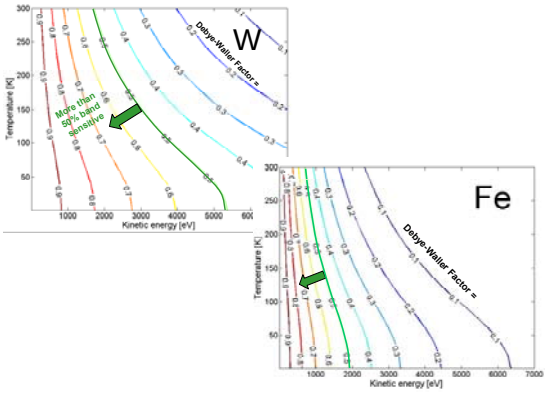




(a) Tiling of detector images. (b) One-step theory. (c) Region in $\vec{k}$-space spanned by images, with associated $\vec{g}_{hk\ell}$ vectors

Debye-Waller (DW) factors permit estimating what might be expected for a number of elemental solids if the photon energy is taken to much higher values up to 10 keV. If we use the rough criterion of at least 50% direct transitions, then for the example of W, one should be able to work at 300K with photon energies up to about 1.7 keV, and with cooling to 4K, with photon energies up to about 5.4 keV. For Fe, the same values are: at 300K up to 0.7 keV and at 4K up to 1.9 keV.

Debye-Waller factors



Fraction direct transitions $\approx W(T) = \exp[-\frac{1}{3} g_{hk\ell}^2 \langle U^2(T) \rangle]$

where g_{hkl} is the magnitude of the bulk reciprocal lattice vector involved in the direct transitions at a given photon energy and $\langle U^2(T) \rangle$ is the three-dimensional mean-squared vibrational displacement.

The Future: ARPES should be possible for many materials in ~1-3 keV range, others up to ≤6 keV (see new data in Papp talk)

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[1] L. Plucinski, J. Minár, B. C. Sell, J. Braun, H. Ebert, C. M. Schneider, and C. S. Fadley, Phys. Rev. B 78, 035108 (2008) and unpublished results.
[2] F. Venturini, J. Minár, J. Braun, H. Ebert, and N. B. Brookes, Phys. Rev. B 77, 045126 (2008) .